PB85-227502

Environmental Radioactivity at the TMI (Three Mile Island), Venting Phase

(U.S.) Environmental Monitoring Systems Lab.
Las Vegas, NV

Jun 85

U.S. Department of Commerce
National Technical Information Service
NTIS.

PB85-227502

EPA/600/4-85/042
June 1985

ENVIRONMENTAL RADIOACTIVITY AT THE TMI, VENTING PHASE

by

E. Bretthauer, F. Grossman, W. Efurd, G. Douglas,
A. Smith, and W. Corkern
Environmental Monitoring Systems Laboratory
U.S. Environmental Protection Agency
Las Vegas, Nevada

and

M. Bills
Office of Monitoring and Technical Support
U.S. Environmental Protection Agency
Washington, D.C.

ENVIRONMENTAL MONITORING SYSTEMS LABORATORY
OFFICE OF RESEARCH AND DEVELOPMENT
U.S. ENVIRONMENTAL PROTECTION AGENCY
LAS VEGAS, NEVADA 89114

REPRODUCED BY
NATIONAL TECHNICAL
INFORMATION SERVICE
U.S. DEPARTMENT OF COMMERCE
SPRINGFIELD, VA. 22161

TECHNICAL REPORT DATA
(Please read Instructions on the reverse before completing)

1. REPORT NO.	2.	3. RECIPIENT'S ACCESSION NO.
EPA/600/4-85/042		PB85 227502/AS

4. TITLE AND SUBTITLE	5. REPORT DATE
ENVIRONMENTAL RADIOACTIVITY AT THE TMI, VENTING PHASE	June 1985
	6. PERFORMING ORGANIZATION CODE: NRD

7. AUTHOR(S)	8. PERFORMING ORGANIZATION REPORT NO.
E. Bretthauer, F. Grossman, W. Efurd, G. Douglas, A. Smith, W. Corkern, M. Bills	

9. PERFORMING ORGANIZATION NAME AND ADDRESS	10. PROGRAM ELEMENT NO.
Environmental Monitoring Systems Laboratory (NRD) PO Box 15027 Las Vegas, NV 89114	XLUF10
	11. CONTRACT/GRANT NO.

12. SPONSORING AGENCY NAME AND ADDRESS	13. TYPE OF REPORT AND PERIOD COVERED
Environmental Monitoring Systems Laboratory-LV, NV Office of Research and Development U.S. Environmental Protection Agency Las Vegas, NV 89114	Response Report 1
	14. SPONSORING AGENCY CODE: EPA/600/07

15. SUPPLEMENTARY NOTES

16. ABSTRACT

From June 28 through July 12, 1980, approximately 44,000 curies of krypton-85 were discharged into the environment from the reactor building housing the crippled nuclear reactor at Three Mile Island (TMI).

The U.S. Environmental Protection Agency (EPA) operated monitoring networks for krypton-85 (5 stations) and airborne particulates (19 stations). These fixed station networks were supplemented, for the duration of the venting, by two mobile units which sampled within the discharge plume.

Calculations of the maximum exposure, based upon measured concentrations of krypton-85, indicated the maximum dose equivalent to the skin to be 0.86 mrem and to the whole body to be 0.0071 mrem. Because the calculated doses derived largely from a few relatively high concentrations of krypton-85, the reported values are well within ±10 percent at the 95 percent confidence level.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group

18. DISTRIBUTION STATEMENT	19. SECURITY CLASS *(This Report)*	21. NO. OF PAGES
Release to Public	Unclassified	14
	20. SECURITY CLASS *(This page)*	22. PRICE
	Unclassified	

EPA Form 2220-1 (Rev. 4-77) PREVIOUS EDITION IS OBSOLETE

NOTICE

This document has been reviewed in accordance with U.S. Environmental Protection Agency policy and approved for publication. Mention of trade names or commercial products does not constitute endorsement or recommendation for use.

ENVIRONMENTAL RADIOACTIVITY AT THE TMI, VENTING PHASE

by

E. Bretthauer, F. Grossman, W. Efurd, G. Douglas,
A. Smith, and W. Corkern
Environmental Monitoring Systems Laboratory
U.S. Environmental Protection Agency
Las Vegas, Nevada

and

M. Bills
Office of Monitoring and Technical Support
U.S. Environmental Protection Agency
Washington, D.C.

SUMMARY

From June 28 through July 12, 1980, approximately 44,000 curies of krypton-85 were discharged into the environment from the reactor building housing the crippled nuclear reactor at Three Mile Island (TMI).

The U.S. Environmental Protection Agency (EPA) operated monitoring networks for krypton-85 (5 stations) and airborne particulates (19 stations). These fixed station networks were supplemented, for the duration of the venting, by two mobile units which sampled within the discharge plume.

Calculations of the maximum exposure, based upon measured concentrations of krypton-85, indicated the maximum dose equivalent to the skin to be 0.86 mrem and to the whole body to be 0.0071 mrem. Because the calculated doses derived largely from a few relatively high concentrations of krypton-85, the reported values are well within ±10 percent at the 95 percent confidence level.

METHODS

Following the reactor accident of March 28, 1979 at the Three Mile Island Nuclear Generating Plant (TMI) the Environmental Protection Agency (EPA) established a long-term routine surveillance program to measure airborne radioactivity. Prior to the krypton-85 purge of the Unit 2 reactor building the routine network consisted of 18 particulate and reactive gas samplers and 2 compressed air samplers (at Goldsboro and the TMI Observation Center) for the collection of noble gases. One particulate and reactive gas sampler was added at Hill Island, and noble gas samplers were added at Middletown, Bainbridge and Hill Island specifically for the purge. In addition, two mobile teams collected samples of particulates and reactive gases, compressed air and atmospheric water vapor during the venting, June 28 to July 12, 1980. Survey meters (PAC-4G with beta probe) were used to attempt to locate the krypton plume, after placement of monitoring personnel based on predictions provided by Lawrence Livermore National Laboratory (LLNL).

Samples for the determination of krypton-85 were collected at the 5 fixed stations and at variable locations within each of 16 (22-1/2°) sectors centered on the Unit 2 Reactor Building as shown in Figure 1. Samples consisted of pressurized tanks of compressed air, each of which contained at least 0.6 standard cubic meters (m^3) of air. The samplers used in the 16 sectors were vehicle mounted (mobile) and generator powered, otherwise they were identical to those at the fixed stations. The fixed station samples were collected and analyzed daily. Mobile samples were taken in each sector only when the winds were predicted to carry the purged gases into that sector. As a result, the number of samples for each sector varied from none to three total samples, the number depending on how frequently the plume was predicted to be in each sector. The sector samples were submitted for analysis as soon as sufficient pressure was attained in each tank. All samples were analyzed by gas chromatography and liquid scintillation techniques (1). The lower limit of detection of this procedure is approximately 4 pCi/m^3; the ambient background concentration of krypton-85 in the area averages approximately 20 pCi/m^3.

Radioactive particulates and reactive gases were sampled by the routine network of 18 surveillance stations (Figure 2) in operation during the purging period plus 1 station added at Hill Island just for the purge. Each sampler utilized a glass-fiber filter to collect particulates followed by an activated charcoal cartridge to collect reactive gases (e.g., iodine-131). The average daily sample represented about 80 m^3 of air. Sample analyses were performed daily by high resolution gamma spectrometry with a lithium-drifted germanium (GeLi) detector and a 10-minute counting time. For this analysis the minimum detectable activity is about 25 pCi for cesium-137 or iodine-131, which corresponds to a minimum detectable concentration of 0.3 pCi/m^3 for an average sample.

The particulate filters were retained, and after the purging was completed one half of each filter, composited by sampling station, was submitted for radiochemical analysis. The composites were analyzed for strontium-89 and -90; uranium-235 and -238; and plutonium-238 and -239 plus -240 by Exxon Nuclear Idaho Company, Inc. Charcoal cartridges were also composited by station and submitted for activation analysis of iodine-129 by Battelle Pacific Northwest Laboratories. Subsequently, the remaining filter portions were halved and submitted to LFE Corporation, Richmond, California for radiochemical analyses as above.

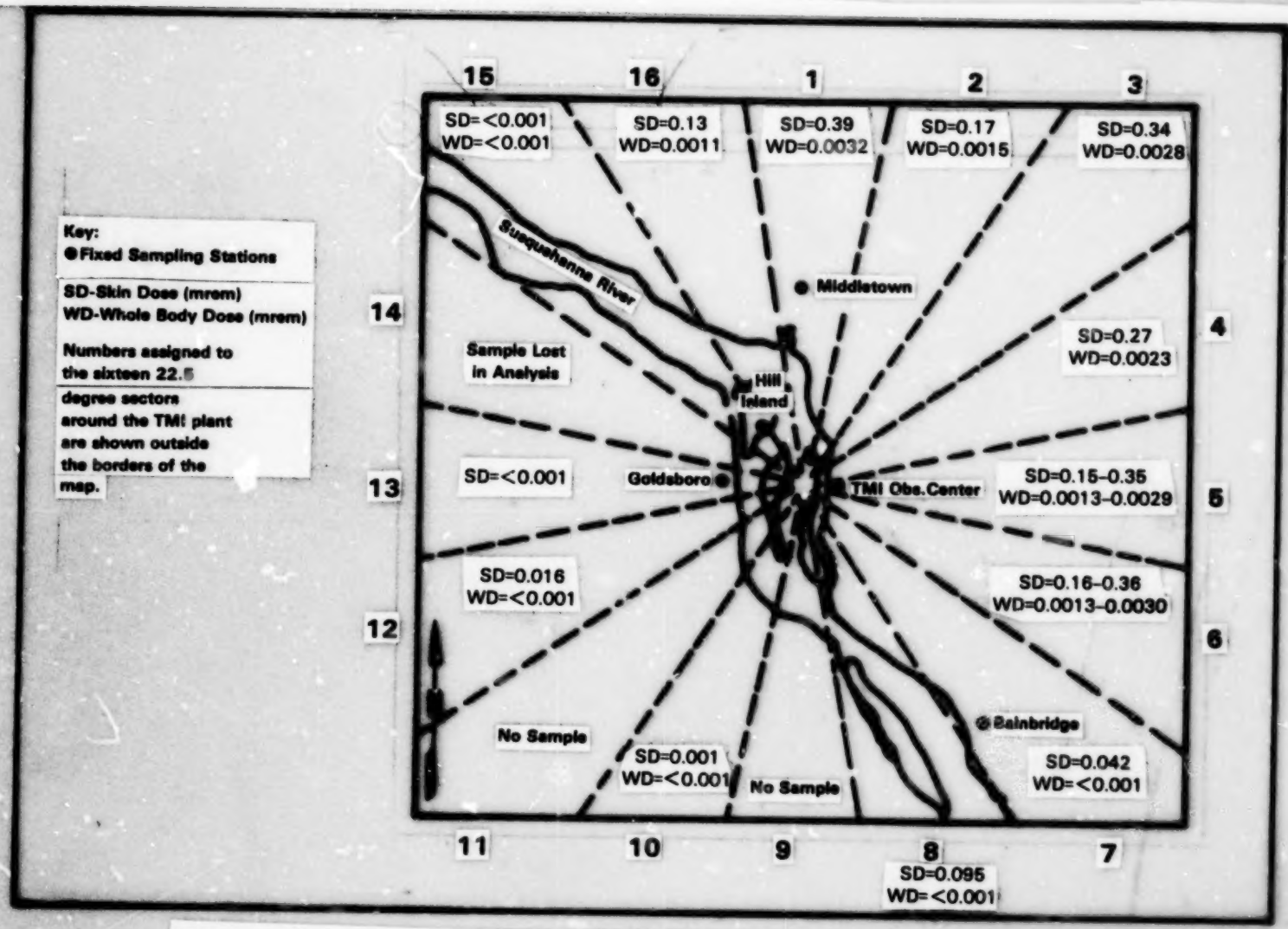

Figure 1. Skin and whole body dose from krypton-85 venting, June 28 through July 12, 1980.

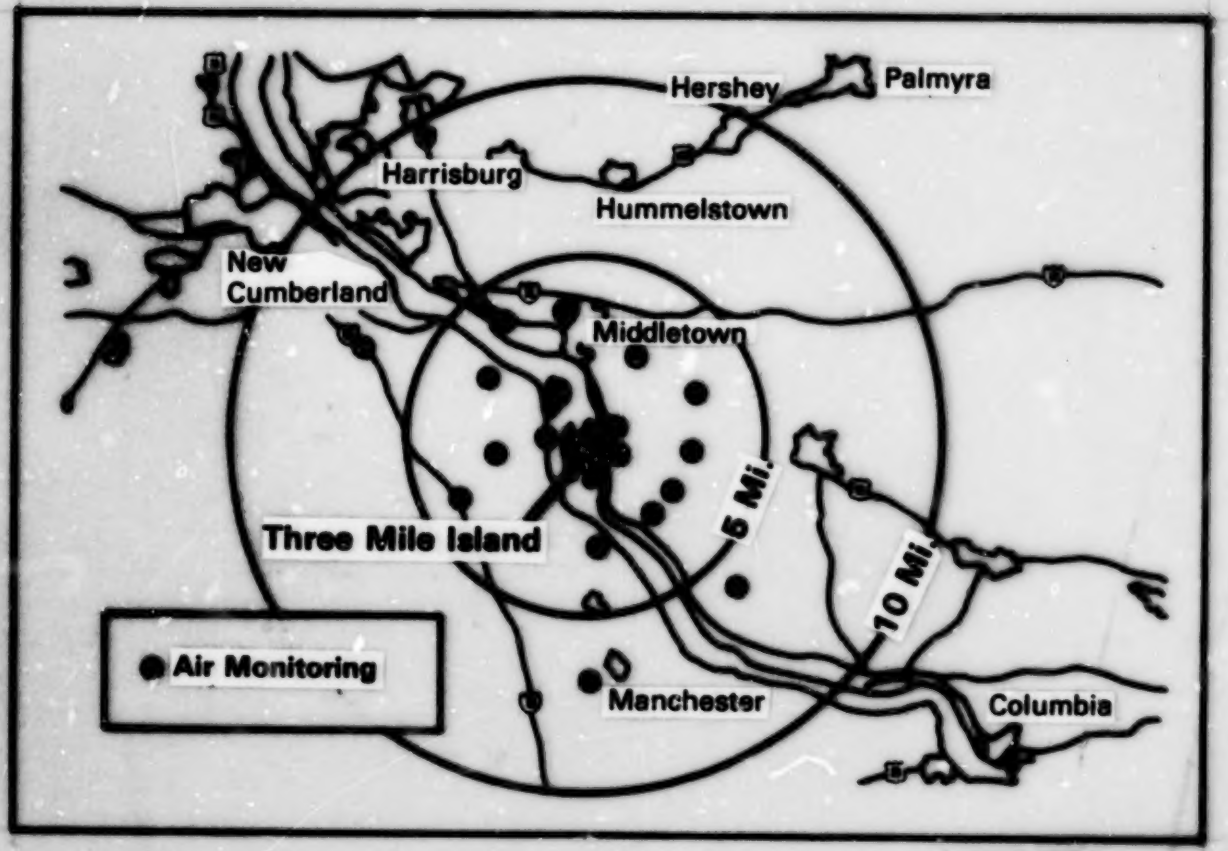

Figure 2. Routine air monitoring network. (Three Mile Island).

A gamma dose-rate recorder was operated at each of the 19 stations, and the recorder charts were examined daily for any increase above background levels.

During the entire period of venting, two mobile monitoring teams comprising personnel from the EPA and the U.S. Public Health Service were operating in the offsite area at all times. Their vehicles were equipped with two-way radios for communication with the EPA control point in Middletown. These teams were positioned at locations where the highest concentration of krypton-85 was expected to occur, as predicted by the U.S. Department of Energy's Atmospheric Release Advisory Capability (ARAC) developed and operated by the Lawrence Livermore National Laboratory (LLNL).

The ARAC is a real-time, computerized atmospheric dispersion model that used wind speed and direction data obtained from weather balloons released from Harrisburg International Airport and all available terrain and surface meteorologic data within 30 kilometers (18.5 miles) of TMI. The model was exercised hourly by personnel of the LLNL in California.

During the first several days of venting, the U.S. Department of Energy's Aerial Measurements System, flown in a helicopter, obtained independent measurements of krypton-85, where it could be detected, to confirm the accuracy of the ARAC predictions.

Once positioned by the ARAC, the monitoring teams utilized hand-held gas-proportional survey instruments capable of detecting krypton-85 concentrations at about one-tenth of the Concentration Guide to refine their position within the plume. The Concentration Guide (300,000 pCi per cubic meter of air) is specified in the Code of Federal Regulations, Title 10, Part 20, as applicable for concentrations of krypton-85, measured at the site boundary, for continuous release to an unrestricted area. Survey meter readings were taken at 15-minute intervals if only background levels were being measured and at 5-minute intervals or more frequently if readings above background were obtained. All readings and times were recorded in a log.

Each vehicle was equipped with a gasoline-powered generator, a compressed air sampler, an atmospheric moisture sampler to enable measurement of tritium as HTO, and an air sampler similar to those in use at the 19 fixed stations. The tritium sampler and standard air sampler were operated concurrently with the compressed air sampler; however, they were operated without regard to sector, whereas the compressed air samples were collected in separate tanks, specific to each sector.

The atmospheric moisture samples were analyzed for tritium by liquid scintillation techniques. The sensitivity of the procedure is 0.4 pCi/ml of water. The particulate filters and charcoal cartridges were analyzed in the same manner as those from the routine stations.

RESULTS

Purging of the reactor building was allowed under a June 12, 1980 Nuclear Regulatory Commission (NRC) Order. For the period of the purge only, a "Temporary Modification of License" was issued, which imposed the following conditions:

> "Do not exceed for the maximally exposed individual in any one of the 16 (22-1/2°) sectors centered on the TMI-2 Reactor Building any of the following:
>
> (a) 15 mrem skin dose
> (b) 5 mrem total body dose
> (c) 20% of the limits in (a) and (b) shall not be over any 1-hour period."

The daily results of the krypton-85 analyses on samples collected at each of the 5 fixed sampling locations are shown in Table 1. The radiation dose equivalent in mrem to the skin and to the whole body of a hypothetical receptor at each location was calculated from these concentrations and the dose conversion factors defined in NCRP Report 44 (2). These dose equivalents for the skin and total body exposures were then summarized for each location and compared to the NRC criteria. As shown by Table 1, the maximum calculated dose equivalent to the skin was 0.86 mrem (<6% of NRC criteria), at the Observation Center, which is at an azimuth of 95° and approximately 0.5 mile from the release point.

The data from the mobile sampling performed in each of the 22-1/2° sectors are presented in Figure 1 as mrem to the skin and to the total body. The values of sectors 5 and 6 are listed as a range because one sample was inadvertently collected across the sector boundary. The value for that sample is assigned totally to each sector to establish the upper limit. The lower limit is defined by deleting that sample from each sector. A similar problem occurred on June 30 - July 1 in Sectors 7 and 8. The skin dose represented by this sample was only 0.002 mrem so the average value is presented in Figure 1.

Gamma spectral analysis was performed on the daily samples (air filter plus charcoal) from the routine monitoring network to confirm the effectiveness of the plant's filtration system for the removal of all radionuclides, except the noble gases and tritium. No fission product radionuclides were detected in concentrations above the minimum detectable level (e.g., 0.3 pCi/m^3 for cesium-134 or -137 for an average daily sample of approximatley 80 m^3).

Overall evaluation of the preliminary results from the initial (Exxon) analyis of strontium-89 and -90, uranium-235 and -238, and plutonium-238 and -239 plus -240 suggested that none of these radionuclides were present in levels greater than those measured in the area prior to venting. However, because several of these results appeared to be positive anomalies, a second portion of every filter was submitted to another laboratory for confirmatory analyses. The second set of results was much like the first; that is, there was a seemingly random distribution of very low positive results (results which exceeded the sample and/or reagent blanks). The qualitative statement, "All

TABLE 1. CONCENTRATION (pCi/m^3) AND CALCULATED DOSE FROM KRYPTON-85

Date	Location				
	Bainbridge	Goldsboro	Hill Island	Middletown	Obs. Center
Daily Concentrations:					
6/27-29	26	34	1400	30	470
29-30	33	*	92	2400	230
30- 1	14	170	560	2300	4300
7/01- 2	34	24	41	18	3500
2- 3	30	97	270	3000	790
3- 4	1400	2400	2400	3300	67000
4- 5	480	760	32	3600	30
5- 6	84	48	1800	340	510
6- 7	36	55	300	160	6900
7- 8	34	220	1200	410	9300
8- 9	34	22	71	420	85000
9-10	420	21	31	42	4300
10-11	23	24	200	29	200
11-12	26	26	19	21	42
Total dose:					
mrem (skin)	0.014	0.019	0.049	0.079	0.86
mrem (total body)	0.00012	0.00016	0.00041	0.00066	0.0071

* No sample collected.

samples show very low or no activity of any of the required nuclides," accompanied this second data set. The few "positive" results should probably be discounted for the following reasons:

1. Only one result from one laboratory indicated the presence of strontium-90 which was, after the cesium isotopes, the most abundant radionuclide in containment. This result was not substantiated by the second laboratory.

2. The ratio of uranium-235 to plutonium-239, -240 is generally an order of magnitude too high to have originated from the reactor.

3. No cesium-137 or cesium-134 was detected on any of the filter samples by gamma spectrometry.

4. The results of the iodine-129 analyses of the charcoal cartridges could not support the consideration that particulate or reactive gaseous radioactivity was released during the venting.

It would, thus, appear that no radioactivity other than krypton-85 and tritium was released during the venting.

Similarly, evaluations of the charts from the gamma-rate recorders at each of the air monitoring stations and of the TLD's deployed during the purge period showed gamma exposure levels within the normal background range.

TABLE 2. RESULTS OF TRITIUM ANALYSES ON ATMOSPHERIC MOISTURE SAMPLES

Mobile Team No.	Collection Period		Tritium Concentration		Water Vapor
	date/time/on	date/time/off	(pCi/m^3 air)	TU($\times 10^2$)	(ml/m^3)
1	6/28 0730	7/6 1015	2.5 ! 1.3	2.1 ! 1.2	3.5
2	6/28 0730	7/6 1000	1.9 ! 0.6	4.0 ! 1.2	1.5
1	7/6 1020	7/11 1500	3.9 ! 2.1	2.9 ! 1.5	4.2
2	7/6 1005	7/11 1400	8.4 ! 2.6	4.0 ! 1.2	6.5

The results of the tritium analyses performed on the atmospheric moisture samples collected by the mobile samplers are listed in Table 2 as Tritium Units (TU) and pCi of tritium per m^3 of air. The amount of water vapor (ml per m^3) recovered from the molecular sieve collectors was inconsistent and lower than would be expected from the relative humidity. Any systematic bias would have been toward lower recovery of the water vapor, due to incomplete removal from the sieves by the laboratory techniques employed in the field. A comparison of the data as expressed in Tritium Units with 1978 and 1979 data reported for atmospheric water vapor by Ostlund and Mason (3,4) indicates that the concentrations determined at TMI were slightly higher than concentrations measured elsewhere. Ostlund and Mason suggest that tritium concentrations at Miami, Florida are indicative of latitudes from 5° to 45° north and that those at Fairbanks, Alaska are indicative of concentrations from 45° to 90° north latitude. The TMI site lies at approximately 40° N so the ambient tritium concentrations would be expected to lie within the range of the Miami and Fairbanks data. Tritium concentrations in water vapor, measured during the summer months ranged from 4.0 ! 0.2 to 9.4 ! 0.7 TU at Miami and from 55 ! 5 to 163 ! 3.6 TU at Fairbanks. Thus, the data from TMI indicate tritium concentrations somewhat above the expected ambient background, although the radiation dose equivalent to the critical organ, the total body fluids, was insignificant, <0.001 mrem.

DISCUSSION

Monitoring for krypton-85 was conducted at 5 fixed locations and by mobile sampling in 16 individual sectors around TMI.

Siting of the fixed stations was based upon:

(a) The prevailing meteorology.
(b) Location of close-in population centers.
(c) Availability of power.
(d) Desirability of locating the sampler at a public building within the population center.

As a result, fixed stations were not necessarily placed at the locations closest to TMI within their sector. The release height of approximately 50 m suggests that the ground-level point of maximum concentration would lie within 1 km under all meteorological conditions encountered during the purge. Only the fixed stations at the Observation Center and on Hill Island were within 1 km of the release point.

The mobile sampling within each sector was designed to provide data at locations other than those covered by the fixed stations. It was our objective to place these mobile samplers at the point of highest predicted concentration of radiokrypton based upon hourly updates of the ARAC predictive model. We expected that, in general, sector samples would show higher concentrations of krypton and higher integrated exposures than the fixed stations because sampling was conducted only during periods of predicted exposure.

In three of the five sectors where fixed stations were located, this is demonstrated. Within the other two sectors, however, the reverse occurred. The data from the fixed station at the TMI Observation Center in sector 5 produced calculated exposures that were significantly (a factor of 2 or more) higher than those calculated from the corresponding sector samples. Because the mobile teams relocated as wind direction changed, the sampling did not pick up residual radioactivity, or that which might have resulted from widespread dispersion of the plume under changing or low windspeed conditions.

Examination of the daily results from the fixed station shows the major contribution to the calculated exposure at the TMI Observation Center occurred on July 3-4 and July 8-9. Of the total radioactivity measured at this site, 37% was collected during the 24 hours from the morning of July 3 to July 4, and 47% was collected from July 8 to July 9. The sector sample that included the July 3-4 interval was the one that was collected in both sector 5 and sector 6. The mobile sampler was not located in sector 5 (at the Observation Center) until 1930 hours on July 3, and sampling was terminated at 0650 hours on July 4. The distance between the sampling location in sector 5 and that in sector 6 is less than 0.25 miles.

Because the concentration of krypton in the sample collected in both sector 5 and 6 was less than (about 1/2) that at the fixed station for the date, it would appear that the mobile team was located at the point of maximum krypton concentration for only a portion of the period of cloud passage.

This sort of widespread plume dispersion could account for most of the differences in integrated exposure. The fixed station exposure calculated at Goldsboro resulted from the measurement of low concentrations of krypton (2400 and 760 pCi/m^3 on July 3-4 and 4-5 respectively). No sector sample was collected during that time interval. A sample collected on June 28 showed only background levels of krypton-85. No other sampling was conducted in that sector, thus there was no integrated exposure calculated from the mobile sampler.

CONCLUSION

The radiation exposure criteria established by the NRC for the purge of the reactor building were designed to assure that no observable health effects would be expected as a result of the release of krypton-85 to the atmosphere.

The data presented demonstrate that the total radiation exposure to the general public was well within the design criteria for the maximally exposed individual (non-occupational). The maximum calculated dose equivalent from krypton-85 was 0.86 mrem to the skin and 0.0071 mrem to the total body. These values correspond to 6% of the skin dose criterion and 1% of the total body dose criterion employed for the purge. Additionally, adherence to the limit of 20% of the dose criteria in any 1 hour was ascertained by field measurements taken with gas proportional survey meters with beta probes and substantiated by the fact that the total dose equivalent was less than 20% of the design criteria.

Although slightly elevated tritium levels in atmospheric moisture were measured during the purge, the incremental dose equivalent to the total body fluids was negligible (less than 0.001 mrem) under the most conservative assumptions.

Essentially all of the gamma-emitting radioactivity in containment as particulates or reactive gases was effectively removed by the exhaust filtration system as demonstrated by the absence of detectable radionuclides on all samples from the air monitoring network.

REFERENCES

1. F. B. Johns, P. B. Hahn, D. J. Thome, E. W. Bretthauer, Radio-chemical Analytical Procedures for Analysis of Environmental Samples (EMSL-LV 0539-17, National Technical Information Service, Springfield, Virginia, March 1979.)

2. Krypton-85 in the Atmosphere-Accumulation, Biological Significance, and Control Technology. (NCRP Report No. 44, National Council on Radiation Protection and Measurements, Washington, D.C., July 1, 1975.)

3. H. G. Ostlund, A. S. Mason, Atmospheric Tritium (Progress Report to Department of Energy under Contract DE-AS05-76EV03944, Rosenstiel School of Marine and Atmospheric Science, University of Miami, Miami, Florida, July 1, 1978 to June 30, 1979.)

4. H. G. Ostlund, A. S. Mason, Atmospheric Tritium (Progress Report to Department of Energy under Contract DE-AS05-76EV03944, Rosenstiel School of Marine and Atmospheric Science, University of Miami, Miami, Florida, July 1, 1979 to June 30, 1980).